

# Effect of Disorder Induced by Heavy-Ion Irradiation on CeCoIn<sub>5</sub> Superconductivity

J.S. Kim · D. Bedorf · G.R. Stewart

Received: 19 May 2009 / Accepted: 29 July 2009 / Published online: 19 August 2009  
© Springer Science+Business Media, LLC 2009

**Abstract** We present a study of the effect of heavy-ion irradiation on a thinned single crystal of the unconventional heavy fermion superconductor CeCoIn<sub>5</sub>. Magnetic susceptibility and low-temperature specific heat results show that the superconducting transition temperature ( $T_c$ ) changed only 4% with irradiation of  $1 \times 10^{12}$  ions/cm<sup>2</sup>, Energy = 1 GeV, while the specific heat jump at  $T_c$ ,  $\Delta C$ , divided by  $C_n$ , where  $C_n$  is the normal state specific heat just above  $T_c$ , was reduced to 3.6 from  $\Delta C/C_n = 4.5$  for unirradiated CeCoIn<sub>5</sub>. The increase of low-temperature magnetic susceptibility and the saturation in magnetization suggest that the defects induced by heavy-ion irradiation are magnetic in nature, as was seen in the case of neutron irradiation on the heavy fermion superconductors, UBe<sub>13</sub> and UPt<sub>3</sub>. The non-Fermi liquid behavior of the irradiated sample, based on the temperature dependence of the low temperature magnetic susceptibility, is significantly altered.

**Keywords** Superconductivity · Heavy fermion · Irradiation · Disorder

## 1 Introduction

A new class of tetragonal heavy-fermion superconductors CeTIn<sub>5</sub> (T = Rh, Ir and Co) has been discovered and attracted much attention in recent years. CeIrIn<sub>5</sub> [1] and CeCoIn<sub>5</sub> [2] show superconductivity at 0.4 and 2.3 K, respectively, while CeRhIn<sub>5</sub>, a heavy-fermion antiferromagnet with  $T_N = 3.8$  K, shows pressure-induced superconductivity [3]. These heavy fermion compounds have been studied intensively.

---

J.S. Kim · G.R. Stewart (✉)

Department of Physics, University of Florida, Gainesville, FL 32611-8440, USA  
e-mail: [stewart@phys.ufl.edu](mailto:stewart@phys.ufl.edu)

D. Bedorf

I Physikalisches Institut, Universitaet Göttingen, Göttingen, 37077 Germany

The unusual properties of CeCoIn<sub>5</sub> have been investigated by various experiments such as specific heat [2, 4–6], magnetic susceptibility [5, 7], resistivity [3, 8], thermal conductivity [4], NMR T<sub>1</sub> measurement [9, 10], and de Haas-van Alphen (dHvA) [8, 11]. These results suggest an unconventional superconductivity in this complex compound.

An H-T phase diagram of CeCoIn<sub>5</sub> [2, 5, 7] shows that the upper critical field where its superconductivity disappears is H<sub>c2</sub> = 5.1 T when the field parallel to the c-axis. At the same field, non-Fermi liquid (NFL) behavior has been reported from the results of a logarithmic temperature dependence of  $C/T$  up to about 10 K [6]. This result that a field tuned quantum critical point (QCP) happens coincidentally at the upper critical field indicates that CeCoIn<sub>5</sub> is close to antiferromagnetic criticality. This could be affirmed by, e. g., the magnetic susceptibility,  $\chi$ , which is not constant at low temperatures and can be well expressed [12] by the non-Fermi liquid dependence  $\chi \sim T^{-1+\lambda}$ ,  $\lambda \sim 0.6$ . Also the linear temperature dependence of resistivity in the temperature range of 2.3–10 K [10] proves the proximity to a QCP. Its nearness to a quantum critical point implies that the superconductivity of this compound is closely linked with its magnetism.

Neutron irradiation was used as a technique to introduce disorder in heavy fermion compounds such as CeCu<sub>2</sub>Si<sub>2</sub> [13], UBe<sub>13</sub> [14, 15] and UPt<sub>3</sub> [15]. In the latter two heavy fermion superconductors, neutron irradiation induces disorder on the ordered state as indicated in reducing the superconducting transition temperature ( $T_c$ ) substantially in UBe<sub>13</sub> and UPt<sub>3</sub> upon neutron irradiation by  $1 \times 10^{18}$  neutrons/cm<sup>2</sup>,  $E > 1$  MeV. Also, a substantial decrease in  $\gamma$  in UPt<sub>3</sub> and the accompanying weakening of the antiferromagnetic spin fluctuations were also observed.

As another candidate for irradiation experiments we chose CeCoIn<sub>5</sub> because it shows a substantially large jump in specific heat,  $\Delta C/C_n = 4.5$  [2], compared with the ordinary BCS value of  $\Delta C/C_n = 1.43$ , where  $C_n$  is the normal state specific heat at  $T_c$ . Another reason is that its superconducting transition temperature,  $T_c = 2.3$  K, is the highest among the heavy-fermion superconductors. Thus, it would be interesting to investigate the effect of disorder induced by irradiation on the superconductivity in this complex compound. Heavy ion irradiation was chosen for this study due to its capability of penetrating the thickness of the sample, resulting in a homogeneously distributed disorder.

Heavy ion irradiation is similar to neutron irradiation in that it produces point defects without introducing chemical impurities (i.e. in contrast to doping experiments), but is dissimilar in that the local elastic strains around the point defects are different.

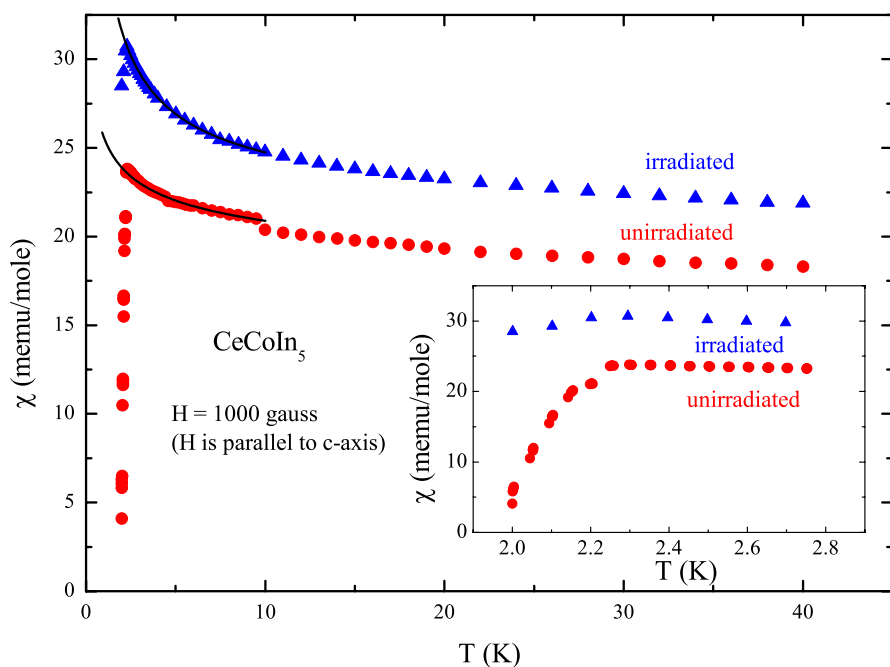
## 2 Experimental

Single crystals of CeCoIn<sub>5</sub> were grown in excess Indium (In) flux with stoichiometric Ce and Co added by following the same procedure used for crystal growth of intermetallic compounds from molten fluxes [16]. The heat treatments followed the sequence as explained in elsewhere [2, 17]. A CeCoIn<sub>5</sub> crystal was polished down to a thickness of  $\sim 16$   $\mu\text{m}$  to insure uniform damage throughout the sample during irradiation. The specific heat and magnetic susceptibility of this polished crystal

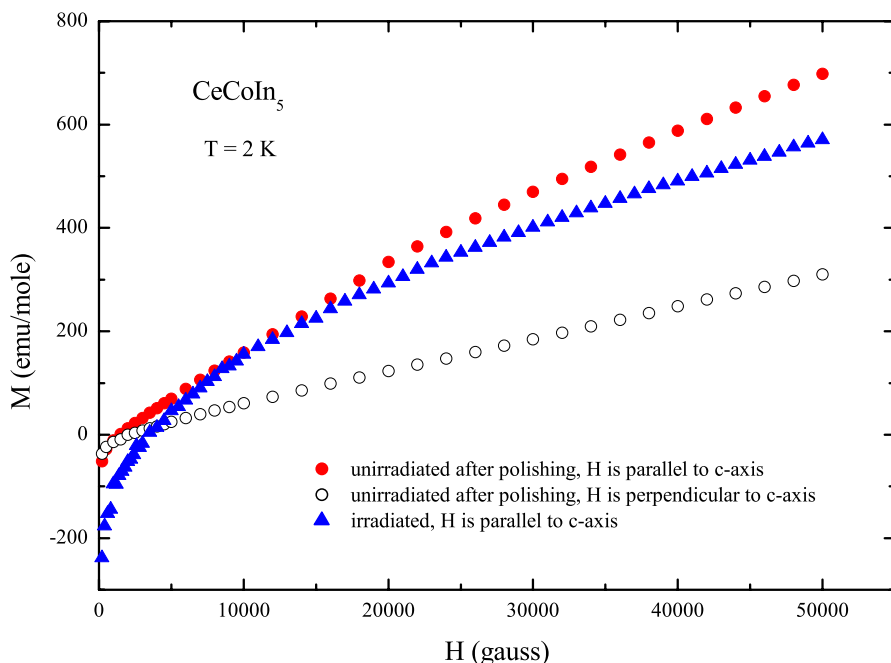
were measured before irradiation to compare with the results after irradiation on the *same* piece of crystal. The irradiation of the  $\text{CeCoIn}_5$  crystalline sample has been done using Pb (lead), at GSI in Darmstadt, Germany. The amount of irradiation was  $1 \times 10^{12}$  ions/cm<sup>2</sup>. The ions were decelerated with Al foil in front of the sample giving an energy of about 1 GeV. Induced radioactivity in the sample and sapphire mounting disk was allowed to decay away before remeasurement, i.e. there is no self-heating problem. The specific heat measurements were made by thermal relaxation method down to 0.4 K [18] and the magnetization in fields up to 5 T and down to 2 K was measured in a commercial SQUID.

### 3 Results

Figure 1 shows the magnetic susceptibility,  $\chi$ , of both unirradiated after polishing and irradiated  $\text{CeCoIn}_5$  measured in  $H = 1000$  gauss applied parallel to the tetragonal c-axis of  $\text{CeCoIn}_5$ . The inset of Fig. 1 is an expanded view of the low-temperature susceptibility to show the onsets of superconducting transition of both unirradi-



**Fig. 1** (Color online) Magnetic susceptibility of single crystal  $\text{CeCoIn}_5$  before and after irradiation with 1 GeV Pb ions at a fluence of  $10^{12}$  ions/cm<sup>2</sup>. Note that the lower temperature limit of the measurements (2 K) is just low enough to see the onset of superconductivity. The black solid lines through the low temperature data are fits discussed in the text to the form  $\chi \sim T^{-1+\lambda}$ , with  $\lambda = 0.4$  for the irradiated sample vs  $\lambda = 0.7$  for the unirradiated sample. The inset shows expanded magnetic susceptibility at low temperatures. Although the data were measured in an applied field of only 1000 G, the magnetization (see Fig. 2) was first measured up to 5 T and there is some trapped flux in the sample causing an effective field of order 3000 G



**Fig. 2** (Color online) Magnetization vs field for single crystal CeCoIn<sub>5</sub> before and after irradiation

ated and irradiated CeCoIn<sub>5</sub>. The susceptibility for both unirradiated and irradiated CeCoIn<sub>5</sub> rises as the temperature decreases until it starts to drop at  $T_c^{\text{onset}} \sim 2.3$  K. The magnetic susceptibility increases more steeply at low temperatures for the irradiated sample. The fits in Fig. 1 shows that the non-Fermi liquid form  $\chi \sim T^{-1+\lambda}$ ,  $\lambda = 0.4$ , for the irradiated CeCoIn<sub>5</sub>, vs  $\lambda = 0.7$  (compare  $\lambda = 0.6$  from [12]) for the unirradiated sample. Thus, irradiation changes significantly the non-Fermi liquid exponent  $\lambda$ . This increased divergent nature of  $\chi$  at low temperatures is only partly responsible for the obvious change in the magnitude of the low temperature data: at  $T = 2.3$  K  $\chi$  of CeCoIn<sub>5</sub> irradiated with  $1 \times 10^{12}$  ions/cm<sup>2</sup> increased 29% after irradiation. This increase is comparable to that seen in UBe<sub>13</sub> (22% increase) and UPt<sub>3</sub> (25% increase) with  $1 \times 10^{18}$  neutrons/cm<sup>2</sup> (see Table 1). Although neutron and heavy ion irradiation produce different concentrations of defects, at least based on the increases in  $\chi$  these relatively small fluences of different disorder-inducing particles seem to have produced comparable levels of change in the low temperature magnetic susceptibility in the three compounds. Meanwhile, the onset temperature of superconductivity for CeCoIn<sub>5</sub> barely changed with irradiation. This result *contrasts* with the neutron irradiation result for UBe<sub>13</sub>, in which  $T_c$  was suppressed 40% from 0.87 K to 0.52 K, and for UPt<sub>3</sub>, in which  $T_c$  was suppressed 60% from 0.5 K to 0.2 K, with a fluence of  $1 \times 10^{18}$  neutrons/cm<sup>2</sup>.

We display the isothermal magnetization curves  $M(H)$  for both directions (parallel and perpendicular to c-axis) for unirradiated CeCoIn<sub>5</sub> to show anisotropy in magnetization and irradiated CeCoIn<sub>5</sub> with field applied parallel to c-axis in Fig. 2. Clearly  $M$  vs  $H$  of irradiated CeCoIn<sub>5</sub> shows more saturation as the field is increased

**Table 1** Comparison with UBe<sub>13</sub> and UPt<sub>3</sub> results [15]. Saturation in  $M$  vs  $H$  was obtained by estimating the difference with the extrapolation of  $M$  vs  $H$  for low fields ( $1\text{ T} < H < 1.5\text{ T}$ )

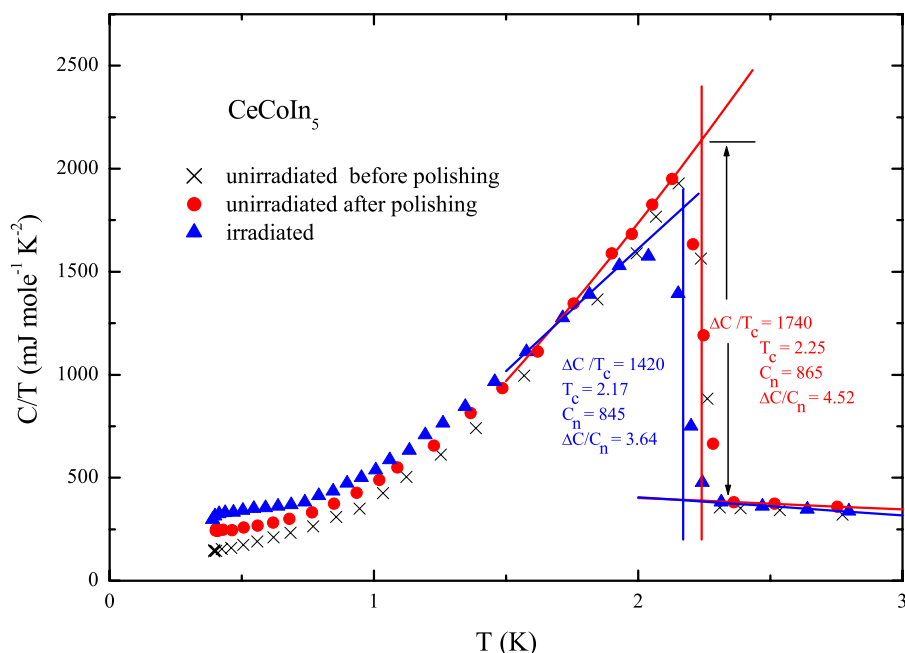
Sample	Irradiation (ions or neutrons/cm <sup>2</sup> )	Low- $T$ $\chi$ (memu/mole)	$T_c$ (K) (onset)	$C_n/T$ (mJ mole <sup>-1</sup> K <sup>-2</sup> )	Saturation in $M$ vs $H$ (%) At 5.5 T
CeCoIn <sub>5</sub>	0	23.8 at $T = 2.3\text{ K}$	2.3 K	385	28
CeCoIn <sub>5</sub>	$1 \times 10^{12}$ (ions/cm <sup>2</sup> )	30.7 at $T = 2.3\text{ K}$ (+29%)	2.2 K (−4.0%)	385 (0%)	41
UBe <sub>13</sub>	0	15.0 at $T = 1.7\text{ K}$	0.87 K	820	6.6
UBe <sub>13</sub>	$1 \times 10^{18}$ (neutrons/cm <sup>2</sup> )	18.3 at $T = 1.7\text{ K}$ (+22%)	0.52 K (−40%)	760 (−7.3%)	7.7
UPt <sub>3</sub>	0	8.0 at $T = 1.7\text{ K}$	0.50 K	440	8.2
UPt <sub>3</sub>	$1 \times 10^{18}$ (neutrons/cm <sup>2</sup> )	10.0 at $T = 1.7\text{ K}$ (+25%)	0.20 K (−60%)	336 (−24%)	14.1

compared to unirradiated CeCoIn<sub>5</sub>, as well as when compared to the magnetization results (Table 1) for neutron irradiated UBe<sub>13</sub> and UPt<sub>3</sub>, where the reported [15] increase with saturation upon irradiation is significantly smaller than observed here for CeCoIn<sub>5</sub>. One of the interesting points to note for the irradiated CeCoIn<sub>5</sub>, the magnetization is negative for fields below  $H = 3000$  gauss at  $T = 2\text{ K}$  while the magnetization remains negative for fields only below  $H = 1000$  gauss at the same 2 K temperature for the unirradiated CeCoIn<sub>5</sub>.

In summary, the increase in low-temperature susceptibilities and increased saturation in  $M$  vs  $H$  in irradiated CeCoIn<sub>5</sub> implies that the defects induced by the heavy ion irradiation are at least partly magnetic in their nature, just as found for UBe<sub>13</sub> and UPt<sub>3</sub> irradiated by neutrons.

Figure 3 shows the temperature dependence of the specific heat divided by temperature for CeCoIn<sub>5</sub> measured on the same piece before and after irradiation. The sample was measured for both before polishing and after polishing for irradiation to check whether the polishing introduced any defects to the sample. As seen in Fig. 3, the specific heat results for  $C_n$  and  $\Delta C(T_c)$  before and after polishing agree with each other. However, the residual  $\gamma$  ( $\equiv C/T$  as  $T \rightarrow 0$ ) in the superconducting state already shows an increase upon polishing, followed by a further increase with irradiation. It should be stressed that these data were taken on the same sample, in the same calorimeter and thus are intercomparable to a precision of better than  $\pm 3\%$ .

The specific heat jump,  $\Delta C/C_n = 4.5$ , for the unirradiated sample agrees with the previous results [2]. This ratio,  $\Delta C/C_n$ , decreased by 20% to 3.6 while the superconducting transition temperature is suppressed from 2.25 K to 2.17 K (only 4%). These superconducting transition temperatures were obtained by considering equal area around  $T_c$  in  $C/T$  vs  $T$  plot. The change in  $T_c$  in CeCoIn<sub>5</sub> by heavy ion irradiation is much smaller than UBe<sub>13</sub> and UPt<sub>3</sub> by neutron irradiation even though the change in the magnetic susceptibility of irradiated CeCoIn<sub>5</sub> is similar to that in irradiated UBe<sub>13</sub> and UPt<sub>3</sub> [15]. Another contrast in CeCoIn<sub>5</sub> is that the normal state specific heat also was not changed by irradiation while the normal state specific heat of both UBe<sub>13</sub> and UPt<sub>3</sub> decreased:  $-7.3\%$  and  $-24\%$ , respectively. This invariance



**Fig. 3** (Color online) Low temperature specific heat,  $C$ , divided by temperature,  $T$ , vs temperature for the same sample of irradiated and unirradiated  $\text{CeCoIn}_5$ . Although the normal state specific heat above  $T_c$ ,  $C_n$ , remains approximately unchanged by irradiation, the discontinuity at  $T_c$  in the specific heat,  $\Delta C$ , is significantly reduced by the irradiation

of the normal state in  $\text{CeCoIn}_5$  by irradiation is in sharp contrast with the case of  $\text{UPt}_3$  irradiation that brought [15] a huge change in normal state properties by erasing almost all the low-temperature upturn from spin fluctuations.

## 4 Conclusions

As a speculation, the difference in the robustness against defects of the upturn in the specific heat in  $\text{CeCoIn}_5$  is influenced by the proximity to a quantum critical point, which is lacking in both  $\text{UBe}_{13}$  and  $\text{UPt}_3$ . If we posit that the superconductivity in  $\text{CeCoIn}_5$  is intimately connected to the quantum criticality, this could further explain the difference in the suppression of  $T_c$  with comparable changes in  $\chi$  between  $\text{UBe}_{13}$  and  $\text{UPt}_3$  (40 and 60% respectively) and  $\text{CeCoIn}_5$  (4%).

In summary, the magnetic properties of  $\text{CeCoIn}_5$  irradiated by  $1 \times 10^{12}$  ions/cm<sup>2</sup> were changed as much (based on  $\chi$ ) or more so (based on the saturation in the magnetization) as those of  $\text{UBe}_{13}$  and  $\text{UPt}_3$  irradiated by  $1 \times 10^{18}$  neutrons/cm<sup>2</sup>. Thus, the nature of the defects induced by irradiation in  $\text{CeCoIn}_5$  appear to be at least in part magnetic, just as in the cases of  $\text{UBe}_{13}$  and  $\text{UPt}_3$ . In contrast, these damage-induced magnetic defects influenced the normal state specific heat and  $T_c$  of irradiated  $\text{CeCoIn}_5$  much less in comparison to both irradiated  $\text{UBe}_{13}$  and  $\text{UPt}_3$ , implying a difference in the (unconventional) superconductivities in the three compounds.

**Acknowledgements** Work at the University of Florida was performed under the auspices of the United States Department of Energy, contract no. DE-FG02-86ER45268. The authors thank C. Trautmann from the GSI Helmholtzzentrum für Schwerionenforschung for technical assistance and H. Hofsäss for fruitful discussions.

## References

1. C. Petrovic, R. Movshovich, M. Jaime, P.G. Pagliuso, M.F. Hundley, J.L. Sarrao, Z. Fisk, J.D. Thompson, *Europhys. Lett.* **53**, 354 (2001)
2. C. Petrovic, P.G. Pagliuso, M.F. Hundley, R. Movshovich, J.L. Sarrao, J.D. Thompson, Z. Fisk, P. Monthoux, *J. Phys. Condens. Matter* **13**, L337 (2001)
3. H. Hegger, C. Petrovic, E.G. Moshopoulou, M.F. Hundley, J.L. Sarrao, Z. Fisk, J.D. Thompson, *Phys. Rev. Lett.* **84**, 4986 (2000)
4. R. Movshovich, M. Jaime, J.D. Thompson, C. Petrovic, Z. Fisk, P.G. Paliugo, J.L. Sarrao, *Phys. Rev. Lett.* **86**, 5152 (2001)
5. S. Ikeda, H. Shishido, M. Nakashima, R. Settai, D. Aoki, Y. Haga, H. Harima, Y. Aoki, T. Namiki, H. Sato, Y. Onuki, *J. Phys. Soc. Jpn.* **70**, 2248, 3187 (2001)
6. J.S. Kim, J. Alwood, G.R. Stewart, J.L. Sarrao, J.D. Thompson, *Phys. Rev. B* **64**, 134524 (2001)
7. T. Tayama, A. Harita, T. Sakakibara, Y. Haga, H. Shishido, R. Settai, Y. Onuki, *Phys. Rev. B* **65**, 180504 (2002)
8. H. Shishido, R. Settai, D. Aoki, S. Ikeda, H. Nakawaki, N. Nakamura, T. Iizuka, Y. Inada, K. Sugiyama, T. Takeuchi, K. Kindo, T.C. Kobayashi, Y. Haga, H. Harima, Y. Aoki, T. Namiki, H. Sato, Y. Onuki, *J. Phys. Soc. Jpn.* **71**, 162 (2002)
9. Y. Kohori, Y. Yamato, Y. Iwamoto, T. Kohara, E.D. Bauer, M.B. Maple, J.L. Sarrao, *Phys. Rev. B* **64**, 134526 (2001)
10. N.J. Curro, B. Simovic, P.C. Hammel, P.G. Pagliuso, J.L. Sarrao, J.D. Thompson, G.B. Martins, *Phys. Rev. B* **64**, 180514 (2001)
11. R. Settai, H. Shishido, S. Ikeda, Y. Murakawa, M. Nakashima, D. Aoki, Y. Haga, H. Harima, Y. Onuki, *J. Phys. Condens. Matter* **13**, L627 (2001)
12. G. Stewart, *Rev. Mod. Phys.* **73**, 797 (2001)
13. G. Adrian, H. Adrian, *Europhys. Lett.* **3**, 819 (1987)
14. N.E. Alekseevskii, V.I. Nizhankovskii, V.N. Narozhnyi, E.P. Khlybov, A.V. Mitin, *J. Low Temp. Phys.* **64**, 87 (1986)
15. B. Andraka, M.W. Meisel, J.S. Kim, P. Wölfe, G.R. Stewart, C.L. Snead Jr., A.L. Giorgi, M.S. Wire, *Phys. Rev. B* **38**, 6402 (1988)
16. P.C. Canfield, Z. Fisk, *Philos. Mag. B* **65**, 1117 (1992)
17. E.G. Moshopoulou, Z. Fisk, J.L. Sarrao, J.D. Thomson, *J. Solid State Chem.* **158**, 25 (2001)
18. G.R. Stewart, *Rev. Sci. Instrum.* **54**, 1 (1983)